PROCESS FOR THE MANUFACTURE OF MULTI-COMPONENT SUBSTANCES

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BACKGROUND

The invention relates to a process for the manufacture of glassy, crystalline or glassy-crystalline multi-component systems without recourse to melting.

Among the transparent multi-component substances known, glasses are the most important. Multi-component glasses are manufactured according to prior art methods by melting, i.e., at temperatures far above the transformation range, at viscosities usually ranging from about 10^2 to about 10^3 poises. Not until this range of temperatures and viscosities is reached can the individual components of the mixture (usually oxides) react with one another in the manner necessary for the formation of glass. Sometimes the temperatures required for the attainment of the above-stated viscosity range are so high that they threaten to make the melting process impractical.

Transparent coatings made by known methods may consist of individual oxides or mixed oxides. Such processes start with hydrolyzable compound which is deposited out of a solvent onto the support where it is hydrolyzed and transformed to an oxide layer by raising the temperature. In this manner, SiO_2 and TiO_2 coatings are prepared without passing through the molten phase. In the preparation of mixed-oxide coatings, the process is limited to those elements which individually form oxides that are resistant to their surroundings — i.e., to air, as a rule, with its ordinary moisture content. No method has become known for introducing, say, alkali oxides or alkaline earth oxides into such layers or coatings.

Nevertheless, the introduction of alkali oxides and alkaline earth oxides, which are known to be glass transforming agents, would signify an important technical advantage, since it would diminish the tendency of such layers to devitrify, which is often triggered by components of the support, and it would also lead to a denser composition in such layers, also due to the fact that heating can then be carried up to the transformation range so that the layers can arrange themselves and densify. The individual oxide and mixed oxide layers, however, have their transformation ranges at such high temperatures that as a rule they cannot be heated up to this range because their supporting materials cannot withstand these temperatures.

The preparation of homogeneous mixtures, for example as starting materials for hydrothermal syntheses, is in the prior art. In this procedure hydroxides are precipitated together at certain pH values, but this yields nothing but mixed hydroxides from which definite glasses (glasses of definite composition) cannot be obtained without passing through the molten phase, or else nitrates are calcined all together and the same thing can be said. According to the prior art, nitrates of other metals are added to alcoholic solutions of silicic acid ethyl ester and the solutions are hydrolyzed with water, whereupon SiO_2 precipitrates as a gel. Calcining follows, in order to decompose the nitrates. This method, too, produces nothing but mixtures. The use of other organometallic compounds is described, such as aluminum isopropylate, triethanolamine titanate, and tetrabutyl titanate. The only advantage of these compounds is stated to be that the heating does not need to be as high as it does with the nitrates. Alkali alcoholates and alkaline earth alcoholates are not mentioned. Alkali is added in the form of carbonate or hydroxide before the hydrolysis, and the cations are simply absorbed into the gel, thus preventing separate crystallization. In the processes known hitherto, therefore, mixtures are always produced from which no definite glassy, crystalline or glassy-crystalline oxidic multi-component solids are obtained without passing through the molten phase. In particular, it is not possible by these methods to apply, for example, glassy coatings to substrates.

THE INVENTION

The invention concerns a general process for the preparation of glassy, crystalline or glassy-crystalline, oxidic multi-component substances, which are or may be transparent, without passing through a molten phase, characterized in that alkali compounds and/or alkaline earth metal compounds that are soluble as solutes in organic solvents are dissolved in organic solvents; other metal compounds of groups I B, II B, III, IV, V, VI, VII A or VIII of the periodic system (Mendelyeev) which are soluble as solutes in organic solvents are dissolved in organic solvents; the alkali or alkali earth metal compounds are reacted with the metal compounds of said Groups I B to VIII in organic solution, so that a homogeneous solution containing the reaction product is formed; then solvent is evaporated in the presence of moisture leaving a residue and finally heating the residue to temperatures below the melting point or melting range of the reaction product.

The alkali compounds or alkali earth metal compounds are preferably used as alcoholates or in a form such that they form alcoholates in solution or form alcoholate complexes with one another in solution. Of the said other metal compounds, P_2O_5, As_2O_3 and H_3BO_3 are examples of preferred materials; other preferred compounds are disclosed in the examples, infra. Alkali and alkali earth metal compounds which can be used, as preferred embodiments, are lithium, sodium, potassium, and calcium and barium; other of said other metal compounds, being preferred embodiments, are magnesium, boron, titanium, silicon, phosphorous, aluminum, zirconium, lead, and zinc. As used herein, "alkali earth metal group" includes magnesium, calcium, strontium and barium.

By the process according to the invention, transparent multi-component substances are obtained even at temperatures that are far below the melting temperature of the substance in question. This is due to the fact that, on account of their reactivity, the components form compounds with one another while in the solvent and during the heating that follows. An example of this is the following reaction:

$$Mg(OR)_2 + Al(OR)_3 \rightarrow Mg\,[Al(OR)_5]$$

wherein the soluble magnesium aluminum alcoholate complex compound forms from magnesium alcoholate and aluminum alcoholate in an alcoholic solution (Houben-Weyl "Methoden der organischen Chemie," Vol. VI/2, Part 2, Page 31, Georg Thieme Verlag Stuttgart, 1963). This complex compound is further processed according to the invention (Example 8) to form spinel. Molecular residues are hydrolytically and/or py-